Average and core silver content of ancient-debased coins via neutron diffraction and specific gravity

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Abstract

The measurement of the fineness of debased ancient silver coins has proven to be a very difficult issue, which has been studied for a long time. In this paper this subject is analysed and the various consequences of the silver surface enrichment (SSE) are discussed exploiting the most recent investigations. A new model is proposed for the complex object that is an ancient debased silver coin, based on the silver profiles measured on some sectioned specimens. The model is applied to a sample of 43 coins, mainly Roman victoriati, Cisalpine and Illyrian drachms (from late III to I century B.C.). The coins are investigated in two different ways: neutron diffraction (ND) and specific gravity (SG). The results of the two measurements are combined via the proposed model to provide a more complete numismatic information of the original fineness of the monetary alloy. As a result, a relation between SSE thickness and SG is derived, which, for these coinages, allows to estimate the original alloy silver content from a simple SG measurement; the same method can be used to study other debased coinages, provided that all the procedure (ND and SG) is applied.

Keywords
ancient silver coins, silver surface enrichment, fineness, specific gravity, neutron diffraction

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Conflicts of Interest

The authors declare that they have no conflict of interest.

1. Introduction

An ancient silver coin was not simply a token, like in modern currencies. In fact, ancient monetary systems were based on the intrinsic value of the coins. The silver content was, together with the weight, the parameter which defined the overall value of a coin (in this paper % stands for wt% when it refers to coins fineness). Therefore, the study of the fineness, or equivalently silver content, would give interesting historical information, but it is not easily measured. Among the latest non-destructive techniques developed as archaeometrical tools one can mention mass spectrometry coupled with laser ablation (LA-ICP-MS: Sarah et al. 2007; Sarah 2008; Blet-Lemarquand et al. 2009), X-ray fluorescence (XRF: Walker 1980; Kasztovszky et al. 2005), and particle induced X-ray emission (PIXE: Bugoi et al. 1999; Ager et al. 2013) as techniques for testing the surface composition of ancient coins. In contrast, prompt gamma activation analysis (PGAA: Kasztovszky et al. 2005; Corsi et al. 2015; Corsi 2015), Fast Neutron Activation Analysis (FNAA: Guerra and Barrandon 1998) and all the various neutron analyses (Calliari et al. 2013; Canovaro et al. 2013; Kirfel et al. 2011; Xie et al. 2004; Kockelman et al. 2003) provide results on the overall coin volume. Remarkable, even if not fully non-destructive, is the latest work of Butcher and Ponting (2015).

The work of Walker (1976) encompassed the XRF analysis of about 2000 Roman Republican coins; his interpretation of the XRF data as bulk results, specifically regarding debased series like Victoriati, caused a distorted perception of this subject for nearly 30 years. He found very stable characteristics, with the silver content oscillating in the narrow range of 92-97% for the whole Roman Republican period. Due to this huge amount of data, no other similar studies have been carried out since then. Recently Cowell and Ponting (2000) also studied the victoriatus coinage by AAS, drilling a small portion of silver from the coin core. The results on some 20 coins provided an average of 68% core fineness, well below Walker results. Even if Walker knew the limitations of XRF for debased silver coinages caused by silver surface enrichment (SSE), he was probably not fully aware of its real extent and consequences, even if since the work of Condamin and Picon (1972) the effect of SSE have been quite well understood and quantified. It is here worth recalling the papers of Beck et al. (2004) and Arles and Téreygeol (2011), where the authors investigate the causes of the presence of SSE in debased silver coins. During the preparation of the blanks, copper tends to oxidize and must be removed; if the procedure is enhanced and chemicals are used to remove the oxides, one can achieve a clean blank which is almost pure silver at the surface. This is most likely the procedure adopted in the ancient mints when the debasement was high, as Beck's and other papers have shown. Similar conclusions are reached in the work by Kasztovszky et al. (2005), where XRF and PGAA results obtained on the same coins are compared.

To evaluate the real silver content, or better the original silver content of the alloy which is still present in the core of the coins, one has therefore to use different techniques, such as the previously referred to bulk methods. XRF is problematic most of the times to ascertain bulk fineness, even on sections of coins (Woyteck et al. 2007) and can be profitably used only for minor element investigations or for highly pure silver coinage (>92% fine). Different kind of XRF instruments has been characterized for the analysis of ancient silver coins (Gore and Davis, 2016) and a campaign of measurements with a hand-held XRF has recently been carried out directly into museums (Corsi et al. 2016b).

This paper combines the technique of neutron diffraction (ND) and specific gravity (SG) with the support of a specific model that describes the composite materials in a debased silver coin. The final purpose is to achieve more precise information on the composition of a debased coin. In fact, ND does not provide the composition of the core, related to that of the original alloy, but an average between the core and the SSE outer part. Those two values can be quite different in highly debased coins, and can differ by as much as 30%, as will be shown. By means of two measurements combined with the proposed model, the two important parameters of the *core composition* and *average composition* can be estimated in a fully non-destructive way. Such data have been available so far only by sectioning the coins (see Sect. 3.1). These two parameters will be the main focus of this paper.

2. Materials and methods

2.1 Neutron diffraction

ND technique is a powerful tool for the analysis of metals, especially in archaeometric studies as precious archaeological artefacts can be analysed in air without any sampling or preparation (Kockelman 2000, 2006). Moreover, results obtained with ND concern all the analysed volume of the objects, hence this technique has been selected for this work.

Neutron diffraction measurements have been carried out at the ISIS pulsed neutron source (Rutherford Appleton Laboratory, Chilton-Didcot, UK) with the diffractometer of the Italian Neutron Experimental Station (INES). INES is a multipurpose powder neutron diffractometer (Grazzi et al. 2007), often used for archaeometric applications (Imberti et al. 2008). The entire volume of each coin has been analysed, adapting the dimensions of the neutron beam thanks to a sample aligner and a neutron camera (Durisi et al. 2013). A detailed description of the setup and data analysis procedure used to calculate the results presented here can be found in a previous paper (Corsi et al. 2016a), where other coins have been analysed using the same instrument and method.

2.2 Specific Gravity

SG relies on a quite simple equipment whose main part is a milligram scale; it is fast (one coin per minute can be investigated) and it does not require expert operators. Moreover, it is inherently a bulk measure, since it analyzes the whole coin.
This technique has been profitably applied to gold (Oddy et al. 1974) and platinum (Willey et al. 2004) but has almost no credit for silver in the ancient numismatic field up to date. This is due to previously experienced major drawbacks regarding reproducibility, lack of accuracy (Caley 1949, 1950) and lack of satisfactory comparisons to other means of investigations, especially for debased silver alloys (Prause 2000). All those issues are addressed in a previous paper (Debernardi 2008b), where the setup and the measuring scheme used for this paper are also described. In the following only the information useful for further considerations present in this paper is reported.
SG (measured in g/cm^3) of each coin has been evaluated measuring its weight (W) and volume (V), obtained through the weight of displaced liquid (W_l) and taking into account the liquid density (ρ_l):

$$\boldsymbol{SG = \rho} = W/V = W/W_l\ \rho_l \tag{1}$$

To simplify several equations it is convenient to introduce as a subsidiary quantity: the inverse of the density $\sigma = 1/\rho$. The relative (ε) precision ε_σ in the measure of σ is the sum of the relative precision of the weight of the coin in air and that of the displaced liquid:

$$\varepsilon_\sigma = \frac{\epsilon_s}{W}\left(1 + \frac{\sigma_l}{\sigma}\right) \tag{2}$$

where ϵ_S is the absolute (ϵ) scale precision (g). Therefore the volume measure introduces the most of the error in the SG value (more than 90% of the total error), because it involves a weight nearly ρ times smaller than the coin weight. The error in the silver content implies to know the relation between SG and alloy composition, which for an ideal binary alloy is given by the well known formula:

$$\text{Ag (wt\%)} = \frac{\sigma - \sigma_{Cu}}{\sigma_{Ag} - \sigma_{Cu}} 100\% \tag{3}$$

where σ_{Cu} and σ_{Ag} are related to the densities of copper and silver (1/8.96 and 1/10.5 respectively). Therefore the absolute precision of the silver content ϵ_{Ag} results in:

$$\epsilon_{Ag} = \frac{\sigma\ \varepsilon_\sigma}{\sigma_{Ag} - \sigma_{Cu}} = \frac{\epsilon_s}{W}\frac{\sigma + \sigma_l}{\sigma_{Ag} - \sigma_{Cu}} \cong \frac{\varepsilon_s}{W} 8000\ (\%) \tag{4}$$

where for σ an average value of the copper and silver values has been used and σ_l =1.26 for ethylene.
For this study, a milligram digital scale was used (50g maximum with 1mg precision, see Fig. 1). In order to achieve more reliable and faster measurements a specially designed tray has been used (Debernardi 2008b), so to eliminate its influence in the volume evaluation (Fig. 1a) and to hold the coin vertically, to avoid the problem of air bubbles on the coin surfaces (Fig. 1d). In the procedure, ethyl alcohol 95% (ethylene) is used and an apparatus calibration is performed using a reference blank, made of pure silver and of approximately the same volume as a denarius. The obtained calibration factor takes into account the SG of ethylene and compensates the contribution of the tray for the increase of the fluid level due to the coin. The overall precision of the measurements can be estimated as ±1 mg, mostly due to the tolerance of the scale, which means an error of about 2% on the fineness for a denarius (see eq. 4).
As previously said (eq. 3), the SG of a silver coin is related to its fineness under the basic hypothesis of a dominantly binary AgCu alloy, which is verified by all the detailed studies on silver ancient coins, based on more sophisticated techniques. The inclusion of other metals (e.g. Pb, Au, Fe) has to be attributed to the limits of ancient metallurgy and was not deliberate; the sum of all of them can be estimated as less than 2%, and then can be disregarded in a first approximation.
However, eq. 3 holds true only for an ideal, freshly prepared AgCu alloy, while in an ancient coin one has to account for the greater complexity of ancient monetary silver, and the possible presence of corrosion products on or in coins long buried in the ground, which can contribute to diminish its SG. Additional effects might be the presence of encrustations, oxides, dirt, etc. The most important phenomenon is however SSE, which is addressed in details in the following.

2.3. Samples

The investigated samples have been analyzed by both ND and SG and are listed in Table 1: the catalog number (column A) will identify the coin throughout the paper, while the coin images are shown in appendix. The analysed coins have been selected on purpose from a larger sample in order to explore all detected SG range and the denarii (No. 39 to 43) have been specifically selected to test the model also for good or slightly debased silver (SG>10). In the table each coin is briefly identified in column B by its RRC number (Crawford, 1974) for victoriati (from no. 1 to no. 23), the Pautasso

type (Pautasso 1966) for the Cisalpine drachms (from no. 24 to no. 33), the moneyer for the Illyrian drachms (from no. 34 to no. 38) and RRC number for the denarii (from no. 39 to no. 43). Column C gives the provenance, which is of two types: private collections ("pc", especially needed for the coins to be cut) and Museo Archeologico di Torino, providing either the Fabretti (1881) or the inventory number. Column D and E report weight and SG. The following four columns (F, G, H, I) provide different Ag contents: columns F and H report respectively the average silver inferred by ND technique and the core alloy composition, inferred by the proposed model in combination with the SG value; column G and I report respectively the silver percentages (average and core) obtained by the only SG data and the proposed model (see section 4.3). Column L reports the difference of the core Ag content measured using the combination of ND and SG minus the same value obtained with SG only. Finally, columns M and N provide the estimated original weight of the coins and the corresponding percentage of weight loss.

3. Theory and calculation

3.1. Microstructure of ancient coins as a consequence of SSE and its effects on SG

The microstructure of different typologies of ancient silver coins has been investigated in several publications (see e.g. Zwicker 1993a, 1993b; Breda 2012; Russo 1996; Arles and Téreygeol 2011; Linke and Schreiner 2000). The analysis of microstructures is particularly useful in numismatic studies because it can provide information about minting techniques and procedures (e.g. striking, cooling speed and eventual heating of blanks). However, the feature of interest in this work for debased silver coins is the presence of a silver-enriched layer on the surface. Indeed, these layers (with thicknesses from a few up to hundreds micrometers) is quite common in objects made of a silver-copper alloy, and some papers in the literature have already discussed and dealt with the topic (Condamin and Picon 1972, Beck et al. 2004; O'Dubhgaill and Jones 2009; La Niece 1993; Ager et al. 2013; Moreno-Suárez et al. 2015). In Fig. 2 some examples are provided: this layer exists on several silver-alloy coins, but is particularly apparent on coins with Ag < 80 wt.%. The nature of this surface layer is manifold and can be explained as:

1- segregation during casting or annealing;
2- deliberate chemical depletion treatments with acids (pickling);
3- wearing and corrosion;
4- post-excavation cleaning with diluted acids.

Some publications report archaeo-metallurgy experiments aimed at understanding the enrichment techniques: Beck et al. (2004) confirmed with some modifications the results obtained by Cope (1972), demonstrating that silver enrichment phenomena (of few μm) already occur in the early stages of blanks preparation, due to the preferred copper oxidation at high temperature. Arles and Téreygeol (2011) completed the study comparing medieval blanks found during archaeological excavations at the Royal Mint in La Rochelle (France) and laboratory-recreated samples. After the reproduction of the entire *chaine opératoire* of an ancient medieval mint, the author concludes convincingly that the enrichment may be the consequence of a series of annealing/hammering process on blanks and a final pickling. Nonetheless, the sequence of these operations leads to silvery layers of about 10 μm, while thickness on archaeological objects reach up to 200-300 μm. The nature of these thick layers is therefore still unclear, but probably linked to pickling baths with acid chemicals that lasted longer than those used by Arles and Téreygeol (2011). As a matter of fact such thick enriched layers have now been experimentally observed in several debased coins, cut for a secure and detailed investigation (Moreno-Suárez et al. 2015, samples N10 and N11 therein, halves of samples 2 and 11 of Table 1). In Fig. 2 and in Moreno-Suárez et al. (2015, Figs. 5 and 6) some examples where the effect of copper depletion is apparent are reported; most importantly, they show that the surface layer is composed of the silver phase and voids that were occupied by the copper grains before they had been oxidized and thereby eliminated from the SSE layer (Condamin and Picon 1972). It is to be remarked that such porous layer is beneath a flat silver surface due to the coin strike, and therefore the liquid cannot access these pores during the SG measurements.

Many groups measured the profiles of silver-copper content vs. the transverse coordinates of the coin section; for example the composition profile of a victoriatus (Moreno-Suárez et al. 2015, Fig. 4) and of a denarius of Septimius Severus (Beck et al. 2004, Fig. 2) show very similar profiles, suggesting that the production technique for debased coins had not changed in about 500 years. In fact in both profiles steady values of Ag and Cu are observed in the core, while close to surface, on similar ranges at around 200-300 μm, the silver increases and the copper decreases.

In Table 2 all the coins that have been cut in this work and their compositional profiles are listed. The surface and core composition are provided; the latter to be compared with the core content inferred by the proposed model. An overall good agreement between the destructive measure (column C) and our estimate (column D) can be observed. Some deviations can be explained by the dependence of column C on the selected section, like it has been observed in Woytek et al. (2007). Average silver content, from ND measurement and simple SG extrapolation are also provided in column E and F; the last column demonstrates that minor components are just due to the limits of ancient metallurgy.

In Fig. 3 data from different measurement techniques are shown and compared for sample no.2. In particular it is worth noting that the data from micro-XRF (volume average = 71.6%, evaluated considering that the enrichment of the rim is the same as the surface. This results in an "amplification" factor of (1+t/R), where t and R are the coin thickness and radius respectively; in fact the rim contributes as $2\pi Rt$, while the two surfaces $2\pi R^2$. The overall result matches well with the average silver content from ND (73.1%), and the micro-XRF core value (61.3±2.4%) is in fair agreement with the value of 62.7% inferred from the model (SG + ND measurements).

The data present a clear clustering of the core silver fineness in the interval 60-70% for the victoriati, which is in fair agreement with the measurements by Mancini of forty years ago (Serafin-Petrillo 1976 and Mancini 1984, 1985). This value is consistent with a technical reason: the eutectic point of the AgCu alloy. This corresponds to a 72% silver content and features a minimum melting temperature of about 780° C. However, alloys in the interval 60-80%, where all the debased coins investigated here lay, display melting temperatures below 850° C (to be compared with 1100° and 950° for copper and silver respectively), which means lower furnace temperatures for the blank production.

The previously described phenomena present a serious problem for the conventional buoyancy method, which works with the assumption of an ideal AgCu binary alloy. This is the origin of the bad reputation of this technique in the ancient numismatic field and can be understood by looking at the histogram of Fig. 10a, which refers to a well-known debased coinage (see e.g. Serafin-Petrillo 1976, Walker 1980, Mancini 1984): the Roman victoriati. In fact, the SG of these coins can be well below the expected values, which, for an ideal AgCu binary alloy, must lay in between those of the two metals, 10.5 for Ag and 8.95 for Cu. Not only does Fig. 10a show values of SG that do not fit with the ideal alloy binary model, being most of the pieces in the interval of SG 7-9, but similar results are reported also by other independent sources. See for example two victoriati RRC 121/1 at ANS: 1944.100.79317 (SG=8.73) and 1968.116.6 (SG=8.98). They are marked as "plated" in the museum trays, as many others. Much lower SG values displays some selected quadrigati at The British Museum: BM 1843,0116.82, SG=8.49; R6350, SG=7.05; BM 2002,0102.161, SG=5.54 (we thank Mr. D. Hook for providing these data). Debased silver coins may lay outside the expected interval simply because that interval applies only to the ideal and homogeneous AgCu alloy. That is a fairly good approximation for good silver, but not at all in the case of debased coins, because SSE transforms the binary alloy in a much more complicated object.

3.2. Model for SG of debased coins

The possibility of modelling the SG of debased coins and estimating their fineness will be discussed in the following. To that aim, first in Fig. 3 typical silver profiles are shown in better detail together with a piece-wise approximation (red dashed line). That will be referred as a "complete copper depletion" model, i.e. it is assumed that the coin is composed just by two homogeneous parts:

1) the bulk, in the core, whose Ag wt.% content is denoted by *x*, related to the fineness of the original blanks,

2) the volume fraction *k* of the SSE layer, which is assumed as an alloy of pure silver and voids, i.e. the spaces freed from copper.

The overall volume of the enriched silver layer is included in the *k* parameter, that takes into account both the composition linear gradient (see Fig. 3) and other geometrical features (coin rim, reliefs, different SSH depths, etc.).
The density ρ (see eq. 1) of a SSE coin is a weighted average of the internal (ρ_i, the core-alloy) and external densities (ρ_e, the Ag-void alloy at the periphery of the coin):

$$\boldsymbol{\rho} = (1-k)\rho_i + k\rho_e \tag{5}$$

For the internal alloy (see also section 2.2), the well-known binary alloy equation holds for the inverse of the density $\sigma = 1/\rho$:

$$\sigma_i = x\left(\sigma_{Ag} - \sigma_{Cu}\right) + \sigma_{Cu} = x\Delta\sigma + \sigma_{Cu} \tag{6}$$

where $\Delta\sigma = \sigma_{Ag} - \sigma_{Cu}$.
The depleted material density in the outer part is easily derived by the following equation:

$$\rho_e = \frac{W_e'}{V_e}\frac{W_e}{W_e'} = \rho_i x \tag{7}$$

where W_e' indicates the weight of the original metal, before the copper depletion and W_e is the weight of the silver in the external alloy, i.e. of the SSE fraction of the coin. Eq. 5 can then be rewritten as:

$$\rho\sigma_i = 1 - k(1-x) \tag{8}$$

which relates the measured density to the original alloy parameter *x*.

Although *k* is not known, it can be related to the measured effective silver content ($\overline{\boldsymbol{x}}$) by:

$$\overline{\boldsymbol{x}} = x(1-k) + kx_e = x(1-k) + k \tag{9}$$

where by definition x_e is set to one. Therefore *k* is linked to the original and average fineness by:

$$k = \frac{\overline{\boldsymbol{x}} - x}{1-x} \tag{10}$$

which, inserted into eq. 8 results in:

$$\boldsymbol{\rho}\sigma_i = 1 + x - \overline{\boldsymbol{x}} = \boldsymbol{\rho}(x\Delta\sigma + \sigma_{Cu}) \tag{11}$$

from which the final equation that relates the two measured parameters $\boldsymbol{\rho}$ and $\overline{\boldsymbol{x}}$ is obtained:

$$x = \frac{\overline{\boldsymbol{x}} + \boldsymbol{\rho}\sigma_{Cu} - 1}{1 - \boldsymbol{\rho}\Delta\sigma} \tag{12}$$

This simple formula allows us to write the fineness of the core alloy *x* as a function of the two measured values. Two measurements are needed because there are two unknowns in the problem: the core composition and the SSE volume fraction *k*. The latter can be obtained from eq. 10, once eq. 12 has provided the value of the former.

Using eq. 10, it is also useful to express *x* as a functions of *k*:

$$x = \frac{\boldsymbol{\rho}\sigma_{Cu} + \mathrm{k} - 1}{k - \boldsymbol{\rho}\Delta\sigma} \tag{13}$$

In Fig. 4 the relation between $\boldsymbol{\rho}$ and fineness *x* of a debased alloy (eq. 13) is plotted for different values of the depletion parameter *k*, i.e. the fraction of the coin volume where SSE takes place.
Due to SSE's strong impact on the SG, an ancient debased coin can show very low SG which, for high debasement and depletion, can even be in the range of 5. Therefore, SG values of debased coins are easily found in or below the SG of copper, as demonstrated by the histogram of Fig. 10a.

In the case of sample no.2, with a core fineness of 61.3% (see Fig. 3), *k* can be estimated at around 0.22. The computation accounts also for the effect of the coin rim, which is enriched as the surfaces (see Fig. 2). A *k* around 0.22, according to the model results in an SG of 9, which is close to the measured value of 8.85. The small difference can be easily attributed to the estimated value of *k* (just based on the average of the two measured sections, when it is well known that variations occur over the transverse position of the coins, see e.g. Fig. 2 for some examples) and to the complete depletion model adopted here.

It is worth underlying that high values of *k* lead to low SG, while at the same time the average silver content may result very high, the higher the bigger *k* is. Correspondingly, also the difference between the core and the average silver content becomes larger. This is due to the fact that copper is replaced by voids (SG=0), so that SG decreases strongly while, disappearing the copper, the average silver content increases correspondingly.

Therefore, for highly debased coins, the measurement of the average silver content can be misleading. In fact, a decent average silver content may result from a quite debased alloy (for example, consider in Fig. 4 the case of SG=7 and k=80%: a coinage produced at a fineness of 60% would result to be better than 90%).

4. Results and discussion

4.1. Application of the model to experimental data

In this section the experimental ND and SG data (Table 1) of the 43 coins investigated in this paper are combined in order to extract all the relevant information, i.e. *x* and *k*.
First, in Fig. 5 the various quantities are reported vs. SG: at left the depletion parameter *k* is plotted for the first time on a fairly large sample of debased coins. As expected, there is a direct relation between *k* and SG, because *k* is the cause of the low SG values in debased silver coins. The values of *k* are found to range from 70% for SG close to 7 down to a few percents for SG higher than 10, which sets a limit for good quality silver.

The results for k show a correlation with the SG, which suggests its exploitation as a "calibration factor" to infer the fineness of a debased coin from just the SG measurement alone. The $k(\rho)$ curve is phenomenologically fitted with a logistic function, that is:

$$k(\rho) = (1 + e^{a\rho+b})^{-1} \tag{14}$$

whose parameters a and b can be easily found by best fitting:

$$a\rho + b = log(k^{-1} - 1) \tag{15}$$

The adoption of a logistic function fitting curve has the advantage of varying between 0 and 1, exactly as the quantity in discussion here. In case of figure 5a also a linear fit would have been suited, but this clearly would fail for SG values lower than about 6, values occurred in measurements of Victoriati.

The corresponding result is reported by the black line in Fig. 5a and in Fig. 5b, 5c its effects on the average and core fineness are also reported together.

Comparing Fig. 5b (average Ag content by ND) and Fig. 5c (estimated core fineness) it can be seen that for high SG (low depletion), the measured and original composition are nearly identical, as expected. The problem arises for highly debased silver, where the difference can approach 30%.
Due to SSE, the average silver content, measured by ND, cannot go much below 70%. The plot of the ND fineness vs. SG (Fig. 5b), which shows how SSE produces a minimum on the average Ag content, is very interesting and enlightening. This behavior is best observed by looking at the k-fitted data, but also the experimental data clearly show it.

In Fig. 6 the difference between the ND fineness and the estimated core alloy Ag content are plotted: at left, the difference is reported vs. SG, where the regular trend follows all the preceding discussion; at right, the same quantity is reported vs. the measured ND fineness. Due to the non monotonic behaviour of the ND fineness vs. SG (see Fig. 5b), the maximum difference is found for ND values at around 90%. Accordingly, a big warning should be given for bulk techniques measuring the average Ag content of a coin, because they risk offering optimistic data for the fineness of debased coins (Condamin and Picon 1972). Such techniques should be always paired with SG, so the full information of the alloy can be retrieved with the model presented here.

It is worth to notice that the core Ag content evaluated with the present model for Victoriati, ranging from 59.6 to 81.6% (see Fig. 5c, red dots), is fully comparable with the one obtained in previous works on the same coinage but with destructive techniques, such as (Cowell and Ponting 2000) where it is found ranging from 52 to 83%.

4.2. Weight loss

The model presented in section 4.1 also allows us to estimate the weight loss of a coin. Under the hypothesis that no silver is lost, this can be simply computed by the weight of the leached copper from the SSE volume, that is:

$$\frac{\Delta W}{W} = k\,\frac{V}{W}\rho_{Cu}(1-x) = k\,\frac{\rho_{Cu}}{\rho}(1-x) \tag{16}$$

In this way one can estimate not only the original alloy, but also the original weight of the blanks. The obtained data are shown in Fig. 7; the reduction is negligible for SG higher than 10 but can reach values as high as 30% for low SG specimens.

It is interesting to investigate the effects of the weight reduction of victoriati on the sample of early victoriati from the Museo Archeologico di Torino, whose SG are reported in Fig. 10a; a detailed list of the coins and all their relevant data is reported in Table 3. In Fig. 8 it can be observed that by accounting for the weight reduction induced by SSE, the expected weight distribution for such early victoriati is recovered. In fact the weight standard for such coins is 3.36 g, i.e. 3 *scripula*. The distribution of the actual weights averages to 2.76 g, while the estimated original weights feature an average value of 3.39 g, quite close to the standard value and is peaked in the 3.2-3.5 g interval. Instead, the histogram of the actual weight is misleading and far from the expected standard. Moreover, the original weight distribution is narrower and better corresponds to the tolerances observed for the denarius series (see Debernardi 2008a).

4.3. Use of the calibration curve to evaluate fineness from SG measure only

The use of the "calibration curve" of eq. 14 can now be discussed; parameters a=1.13, b=-9.23 best fit the measured data. In Fig. 5b and 5c the resulting average and core Ag content are shown, which fairly fit the measured data. It is therefore interesting to see in Fig. 9 the difference between the measured and "calibrated" results of the core Ag

composition. The standard deviation amount to 5% and 39 of the 43 specimens lay between ± 6% difference; such differences can be mostly attributed to the approximation of full depletion and to different oxidation processes (see e.g. Fig. 2) that locally modified the density.

There is no apparent difference among the various drachma coinages, which suggests that all those debased coinages have been produced with similar techniques, even if in different places and periods (Victoriati and Cisalpine drachms are about contemporary, while the Illyrian coins are later). Instead, for denarii of much higher silver content and SG (they have been chosen mostly from the period of the Social War, when the coinage was debased), it is worth noting that the difference between the measured and calibrated data is always below ± 0.9% with a standard deviation of 0.7%.

Overall, estimating the fineness of ancient silver coins, debased or not, with the measurement of SG and applying the proposed model has become quite reliable. It provides a maximum error bar of ± 6% for highly debased coins and ± 1% for good silver. The neutron techniques are bulk techniques that provide an average of the sample's volume. However this has the drawbacks discussed above with reference to Fig. 4: in the case of highly debased coins the neutron techniques result in much higher silver contents (as higher as 30%) compared to the core alloy. Therefore one cannot rely on neutron techniques in highly debased coins unless accompanied by an SG measurement.

This is exemplarily illustrated in Fig. 10, where the same early victoriati of Museo Archeologico di Torino, whose weight statistics are reported in Fig. 8, are investigated via SG model only, reporting both the core and average fineness. Comparing the two distributions one can clearly see the relevant effect of the SSE layer on the distributions of the two quantities. The average silver content is not only shifted 12% toward higher silver contents (averages 67 and 79% respectively), but its shape is also modified. The average fineness starts at 75% from where it continuously decreases, while the core fineness presents a peak at 66%, but has also a tail towards lower values.

At the same time, Fig. 8 to Fig. 10 shows that the only SG measure, thanks to the proposed model and to the calibration factor extracted using also ND data, allows to extract all the relevant alloy parameters of a coinage.

5. Conclusions

In this work we have presented a model useful for the calculation of the original fineness of ancient coins, before any blank treatment and before any alteration due to long burials. The method combines two measurements to achieve more detailed information about highly debased coins in a fully non destructive procedure. The advantage over alternative approaches is that it provides volume information, i.e. not bound to specific regions of the coins. In fact, both SG and ND give an average result of full coin volume.
The model has been applied to 43 coins, for which measurements of both specific gravity and neutron diffraction were performed. Some coins have been sectioned, as well, to study their internal microstructure and their silver surface enrichment (SSE) areas. Such detailed investigations demonstrate that the proposed technique is reliable and can be used to study other debased coinages provided that all the procedure (ND and SG) is applied.
The important warning from this research is that, in highly debased coinages, ND results alone do not provide precise information on the fineness of the core alloy: the average of the core and the SSE region might result in overestimating the silver content in the original alloy up to 30%.

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Figure captions

Fig. 1 The SG-meter used in this work and its four measurement steps for the coin volume. a, holder is tared out; b, holder is lifted; c, coin is inserted in vertical position; d, holder is set to its rest position, where "volume" is read out

Fig. 2 optical microscope images of the sections of an Illyrian drachm (no. 34) and of a victoriatus (no. 23); for no. 23, various levels of magnifications are provided

Fig. 3 Ag profiles at two different positions (blue and green dots) vs. depth of the coin (sample no. 2, i.e. sample N10 in Moreno-Suárez et al. 2015). The red-dashed line show the complete depletion model proposed here, while the black line the estimated average from the micro-XRF data. The continuous lines, red and black, indicate the estimated fineness averages inferred from ND and micro-XRF data respectively. Inset: optical microscope image of the sectioned coin (Angelini et al. 2013)

Fig. 4 Core (continuous lines) and average (dashed lines) Ag percentage as a function of SG for different values of *k* (SSE percentage); the circle (square) refer to the measured core (estimated average) *x* vs. measured SG of coin no.2 (see Fig. 3)

Fig. 5 Experimental ND and SG data applied to the model. (a), the estimated volume fraction *k* of SSE vs. SG; (b) the measured fineness average (col. F in Table 1) vs. SG and (c) the corresponding extrapolated values for the original alloy (col H in Table 1). The lines (see text) refer to values resulting from fitting *k* vs. SG with eq. 14: a=1.13, b= -9.23

Fig. 6 Difference between ND Ag percentage (col F in Table 1) and corresponding estimate of the core alloy (col H in Table 1). At left, this quantity is reported vs. SG, at right, vs. the ND data itself

Fig. 7 Weight in the measured sample. At left, weight loss vs. SG, showing also the *k* -fit result. At right, the actual weights (colored circles) are compared with the original weights (black dots) of the blanks before the SSE process and/or during the burial

Fig. 8 Weight histograms of the 30 victoriati sample from Museo di Antichità di Torino. Left, actual weights; right, estimated weights of the original flans. Bin-size is 150mg. The weight of the half victoriatus v23 (see Table 3) is doubled in these diagrams

Fig. 9 Difference between the estimated core Ag content and the one computed using the "calibration curve" of Fig. 5c, i.e. only the SG values are used (see Table 1, col. L; the corresponding standard deviation is 5%)

Fig. 10 a) SG histogram of 30 victoriati of the earliest series in Museo di Antichità di Torino (see Table 3 and also Angelini et al. 2013); b) and c): the corresponding core and average silver distribution computed from SG only via the calibration factor *k* and the model. Bin-sizes: 0.3g/cm^3 for SG, 2.5% for fineness

Appendix Fig. Images of the investigated coins; all details are provided in Table 1

Table captions

Table 1: Data of the investigated coins: see text for a detailed description of the columns.

Table 2. Results from different investigations. All the samples listed here have been cut to investigate the inner structure and composition. Column B reports the surface fineness measured by SEM-EDX or µ-PIXE techniques; C: core fineness by SEM-EDX or µ-PIXE measurements; D, core fineness from model (combined ND and SG); E, average fineness from ND only; F, average fineness from just SG and calibration curve; G, core minor elements weight percentage by SEM-EDX or µ-PIXE measurements.

Table 3: Data of the investigated victoriati of the Museo Archeologico di Torino, with their inventory or Fabretti numbers (Fabretti 1881). The column A is a catalog number for these victoriati. Column B indicates if the coin is comprised in the catalog of Table 1, providing the corresponding catalog number. Museum references and *RRC* type (Crawford, 1974) follow in column C and D. Weight, SG, core and average fineness are reported in columns E, F, G and H respectively. In columns I and L the original flan weight and the corresponding weight loss is reported.

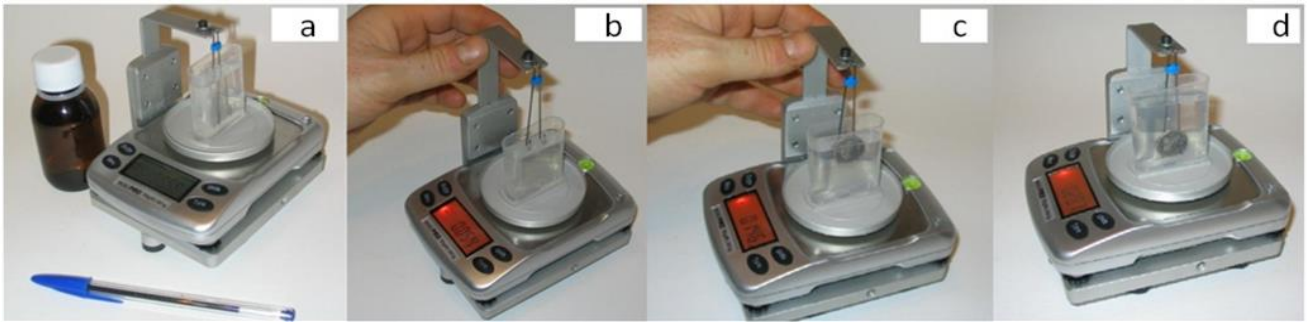

Figure 1

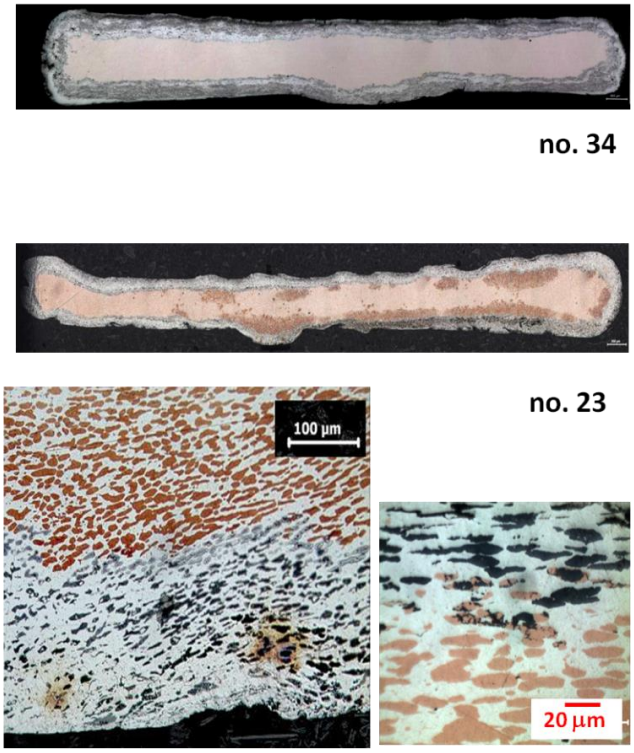

Figure 2

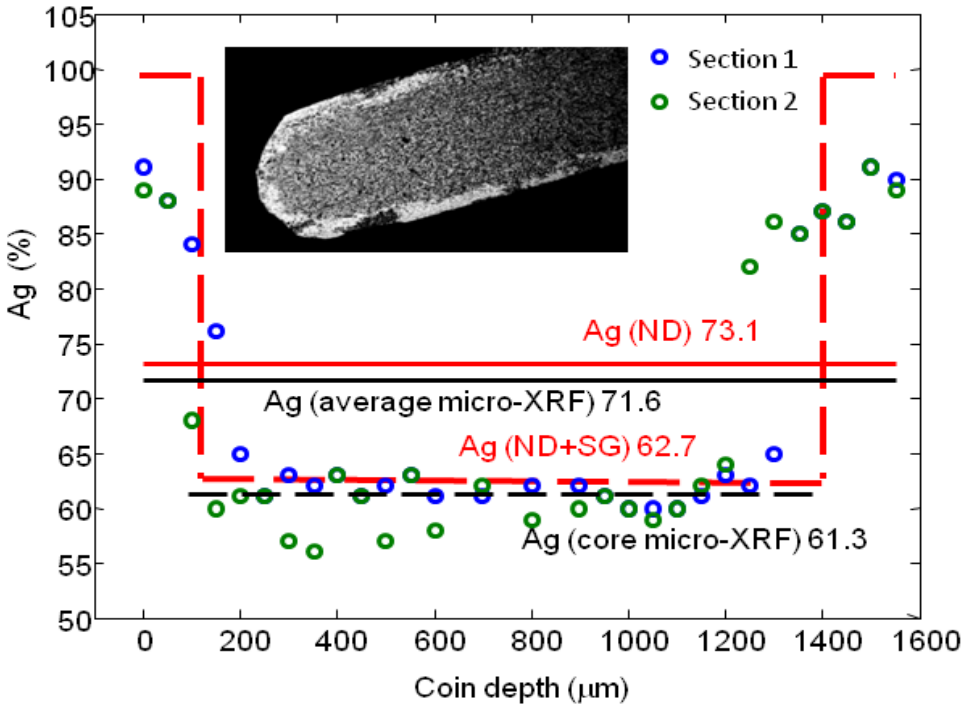

Figure 3

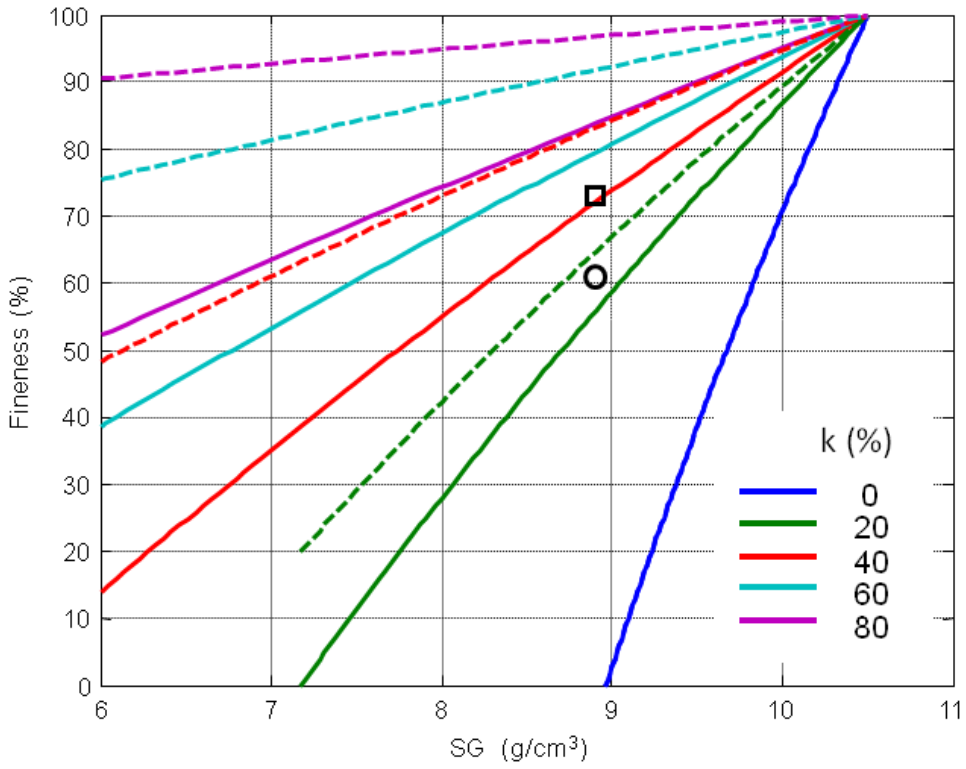

Figure 4

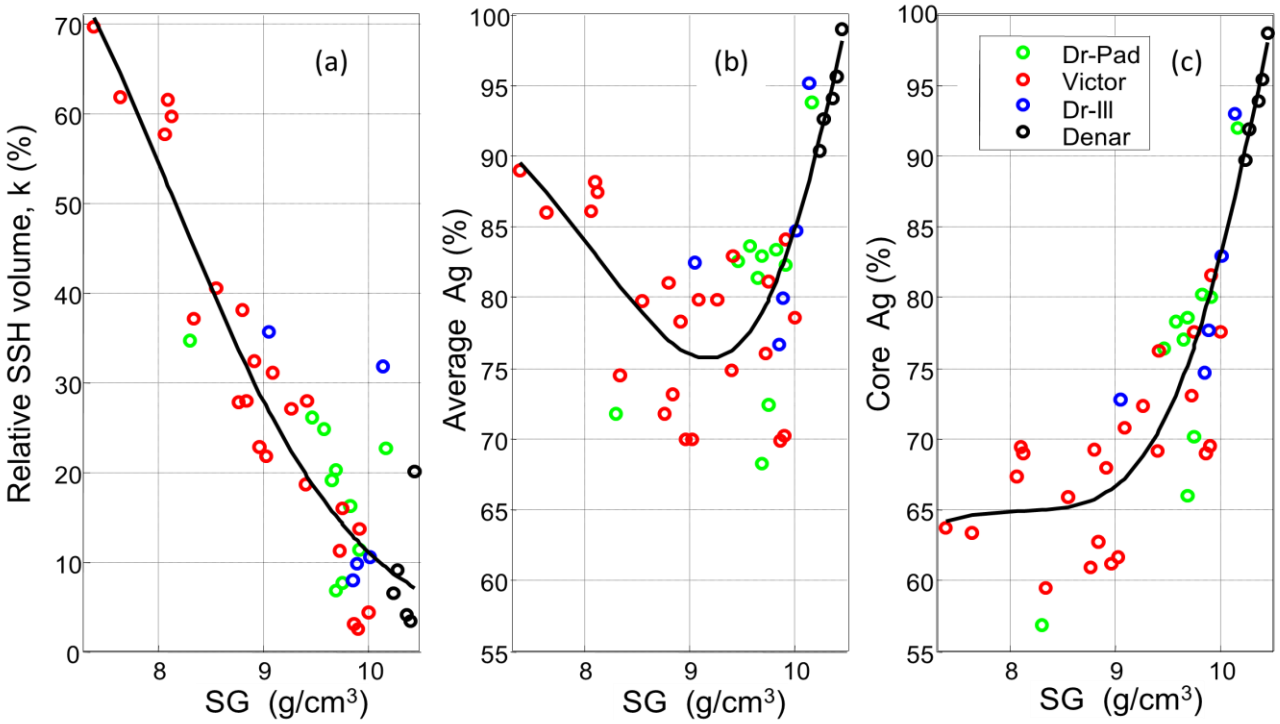

Figure 5

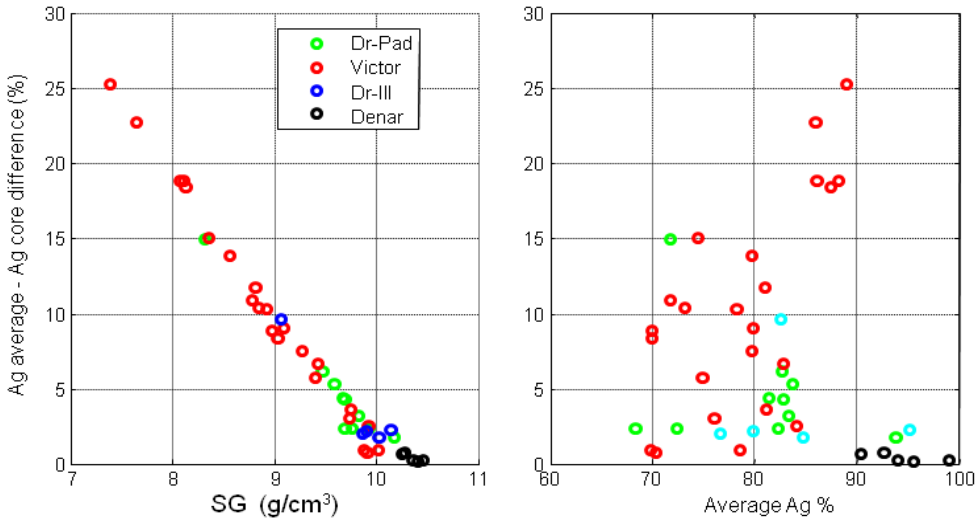

Figure 6

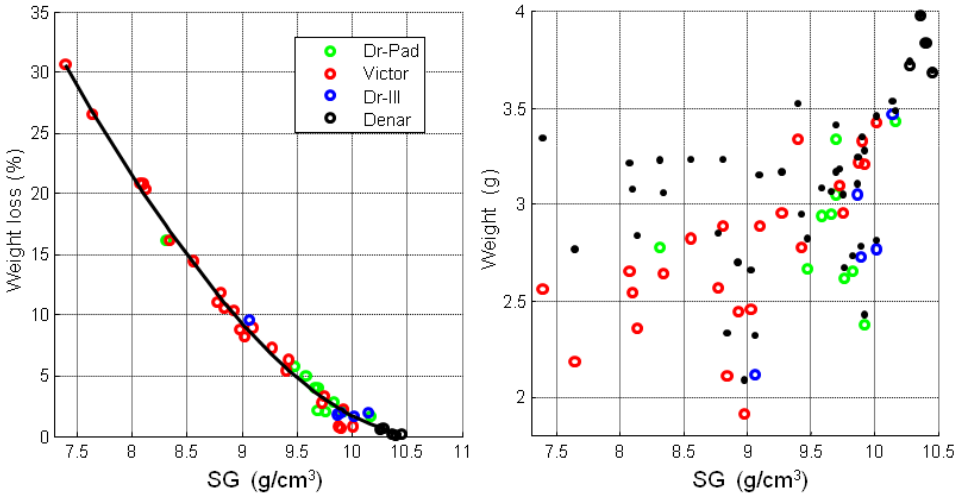

Figure 7

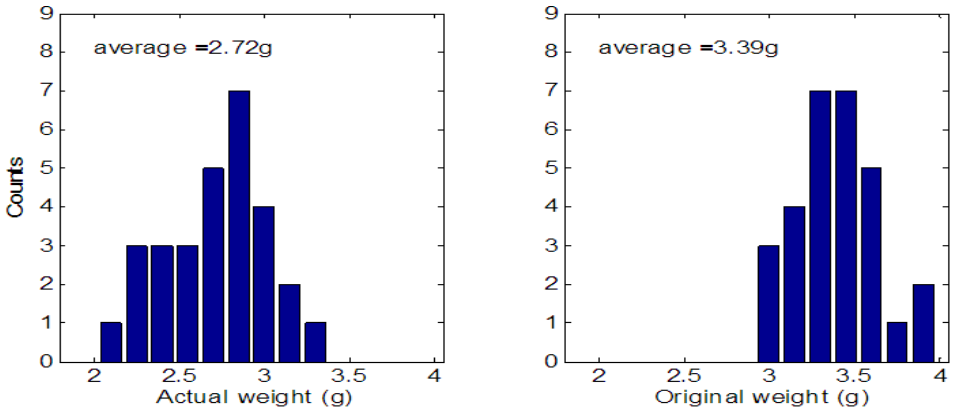

Figure 8

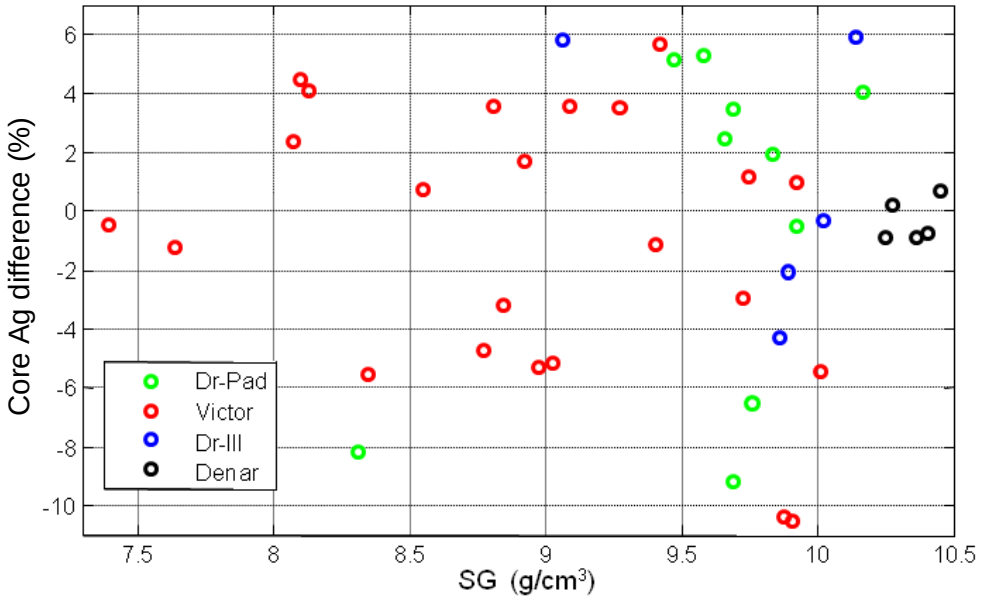

Figure 9

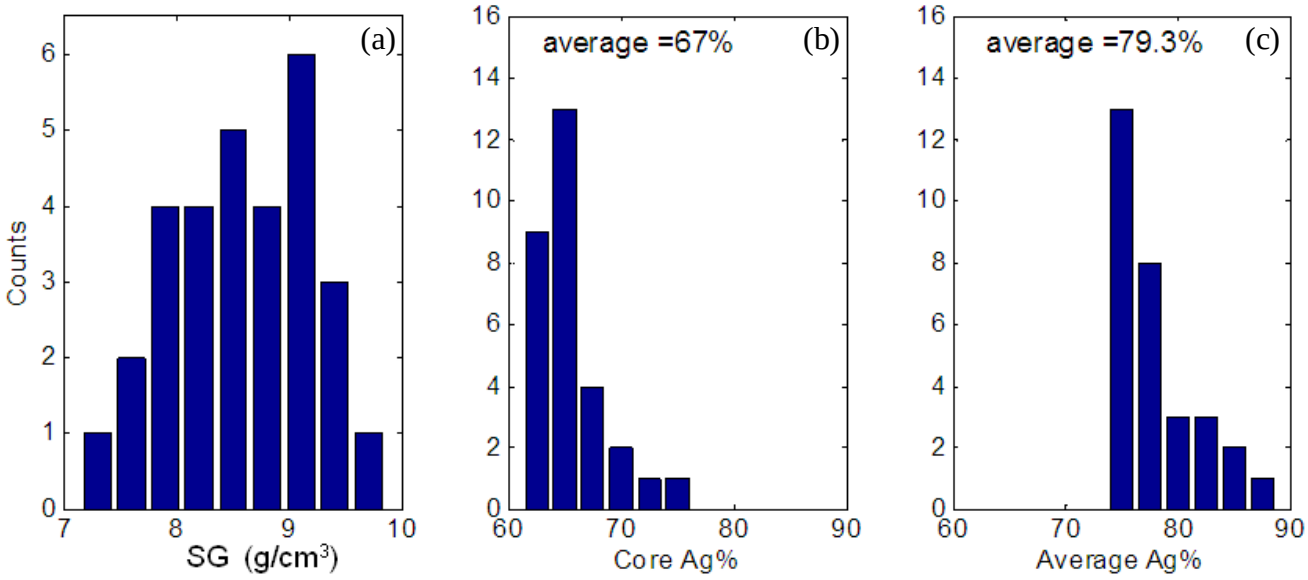

Figure 10

Appendix

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A	B	C	D	E	F	G	H	I	L	M	N
No.	*RRC/type*	*Ref.*	*Actual Wt. (g)*	*SG (g/cm^3)*	*Av. Ag - ND (wt.%)*	*Av. Ag SG only (wt.%)*	*Core Ag ND + SG (wt.%)*	*Core Ag SG only (wt.%)*	*Core Ag difference (wt.%)*	*Orig. Wt. (g)*	*Wt. loss (%)*
1	44/1	pc	3.22	9.87	69.9	82.0	69.0	79.4	-10.4	3.25	1
2	53/1	pc	2.11	8.84	73.1	76.8	62.7	65.9	-3.2	2.33	10
3	53/1	F. 231	2.56	7.39	89.0	89.5	63.8	64.2	-0.5	3.35	31
4	53/1	pc	2.89	8.81	81.0	77.0	69.3	65.8	3.5	3.24	12
5	57/1	pc	2.45	8.92	78.3	76.3	68.0	66.2	1.7	2.70	10
6	70/1 var	pc	3.34	9.40	74.9	76.2	69.2	70.3	-1.2	3.53	5
7	70/1	pc	3.43	10.01	78.6	85.0	77.6	83.1	-5.5	3.46	1
8	70/1	pc	3.21	9.92	84.1	82.9	81.6	80.6	1.0	3.28	2
9	71/1	F. 648	2.55	8.10	88.2	83.1	69.4	64.9	4.5	3.08	21
10	71/1	Inv. 11578	2.78	9.42	82.9	76.3	76.3	70.6	5.7	2.95	6
11	83/1b	pc	2.96	9.27	79.8	75.8	72.3	68.8	3.5	3.17	7
12	89/1b	pc	2.96	9.75	81.1	79.8	77.6	76.4	1.1	3.05	3
13	93/1b	pc	3.10	9.73	76.1	79.5	73.0	76.0	-3.0	3.18	3
14	93/1a	F. 675	2.66	8.07	86.1	83.4	67.3	64.9	2.4	3.22	21
15	93/1a	pc	3.33	9.90	70.3	82.6	69.6	80.1	-10.6	3.35	1
16	95/1a	pc	2.46	9.03	70.0	75.9	61.6	66.8	-5.2	2.66	8
17	98/1a	F. 666	2.89	9.09	79.8	75.8	70.8	67.2	3.5	3.15	9
18	98A/1a	F. 672	2.19	7.64	86.0	87.4	63.4	64.6	-1.3	2.77	26
19	102/1	F. 601	2.83	8.55	79.7	78.9	65.9	65.2	0.7	3.23	14
20	166/1	pc	2.36	8.13	87.5	82.9	69.0	64.9	4.1	2.84	20
21	166/1	pc	2.57	8.77	71.8	77.2	60.9	65.7	-4.7	2.85	11
22	166/1	pc	2.64	8.34	74.5	80.8	59.5	65.0	-5.5	3.06	16
23	166/1	pc	1.92	8.97	70.0	76.1	61.2	66.5	-5.3	2.09	9
24	Naturalistic lion	Inv. 61851	2.28	9.92	82.3	82.9	80.0	80.5	-0.5	2.43	2
25	Wolf lion	Inv. 61843	2.78	8.31	71.7	81.1	56.8	65.0	-8.2	3.23	16
26	Wolf lion	Inv. 61822	2.62	9.76	72.3	80.0	70.1	76.6	-6.5	2.67	2
27	Wolf lion	Inv. 66483	3.34	9.69	68.3	78.9	66.0	75.2	-9.2	3.41	2
28	Scorpion lion	Inv. 87509	2.95	9.66	81.4	78.5	77.0	74.6	2.5	3.07	4
29	Scorpion lion	Inv. 66500	2.94	9.58	83.7	77.6	78.3	73.1	5.3	3.09	5
30	Naturalistic lion	Inv. 87502	2.67	9.47	82.6	76.7	76.4	71.3	5.1	2.83	6
31	Scorpion lion	Inv. 66493	2.66	9.83	83.3	81.1	80.2	78.3	1.9	2.74	3
32	Scorpion lion	Inv. 66503	3.05	9.69	82.9	78.9	78.6	75.2	3.4	3.17	4
33	Massa beta	Inv. 22653	3.43	10.17	93.9	89.1	92.0	87.9	4.0	3.48	2
34	Apollonia, Ariston	pc	2.12	9.06	82.5	75.8	72.8	67.0	5.8	2.32	9
35	Apollonia, Timen	pc	2.73	9.89	79.9	82.3	77.7	79.8	-2.1	2.78	2
36	Dyrrachium, Meniskos	pc	2.77	10.02	84.7	85.1	82.9	83.3	-0.3	2.81	2
37	Apollonia. Ariston	pc	3.05	9.86	76.7	81.7	74.7	79.0	-4.3	3.11	2
38	Illyrian drachm, Imitation	pc	3.47	10.14	95.2	88.3	93.0	87.1	5.9	3.54	2
39	346/1b	pc	3.72	10.28	92.6	92.4	91.9	91.7	0.2	3.74	0.6
40	348/1	pc	4.38	10.25	90.4	91.4	89.7	90.6	-0.9	4.41	0.6
41	348/3	pc	3.98	10.36	94.1	95.1	93.8	94.7	-0.9	3.99	0.2
42	349/1	pc	3.84	10.40	95.6	96.5	95.5	96.2	-0.8	3.84	0.1
43	350B	pc	3.69	10.45	99.0	98.2	98.7	98.0	0.7	3.70	0.2

Table 1: Data of the investigated coins: see text for a detailed description of the columns.

A	B	C	D	E	F	G
No.	*Surface Ag (wt.%)*	*Core Ag (wt.%)*	*Model core Ag (wt.%)*	*Average ND Ag (wt.%)*	*Average model Ag (wt.%)*	*Core minor elements (wt.%)*
2	98.8 ± 0.4	63.7 ± 0.4	62.7	73.1 ± 1.0	76.8	Au 0.17; Fe 0.11; Pb 0.39
11	89.5 ± 0.3	56.7 ± 0.2	72.3	79.8 ± 1.1	75.8	Au 0.20; Fe 0.16; Pb 0.35
23	93.6 ± 0.3	54.6 ± 0.3	61.2	70.2 ± 1.0	76.1	Au 0.33; Pb 0.31
34	85.0 ± 0.3	71.1 ± 0.3	72.8	82.5 ± 1.2	75.8	Au 0.33; Fe 0.09; Pb 1.16
35	95.7 ± 0.4	71.4 ± 0.3	77.7	79.9 ± 1.2	82.3	Au 0.35; Fe 0.03; Pb 1.40
36	93.7 ± 0.4	77.4 ± 0.3	82.9	84.7 ± 1.2	85.1	Au 0.30; Fe 0.04; Pb 1.58

Table 2. Results from different investigations. All the samples listed here have been cut to investigate the inner structure and composition. Column B reports the surface fineness measured by SEM-EDX or µ-PIXE techniques; C: core fineness by SEM-EDX or µ-PIXE measurements; D, core fineness from model (combined ND and SG); E, average fineness from ND only; F, average fineness from just SG and calibration curve; G, core minor elements weight percentage by SEM-EDX or µ-PIXE measurements.

A	B	C	D	E	F	G	H	I	L
num.	*Present cat.*	*Fabretti/Inv.*	*RRC*	*Actual Wt. (g)*	*SG g/cm^3*	*Core Ag (wt.%)*	*Average Ag (wt.%)*	*Orig. Wt. (g)*	*Wt loss %*
v1		Inv.11572	44/1	**2.263**	**7.64**	**65**	**87**	3.37	49
v2		227	44/1	**2.956**	**8.29**	**65**	**81**	3.92	33
v3		233	44/1	**3.320**	**9.68**	**75**	**79**	3.67	11
v4		234	44/1	**3.004**	**8.46**	**65**	**80**	3.87	29
v5		235	44/1	**2.800**	**8.76**	**66**	**77**	3.43	23
v6		228	44/1	**2.886**	**8.70**	**65**	**78**	3.57	14
v7		229	53/1	**2.868**	**9.11**	**67**	**76**	3.35	17
v8	3	231	53/1	**2.562**	**7.39**	**64**	**89**	3.97	55
v9		Inv. 11571	53/1	**2.850**	**9.24**	**68**	**76**	3.28	15
v10		236	70/1	**2.555**	**9.04**	**67**	**76**	3.01	18
v11		647	71/1	**2.529**	**8.26**	**65**	**82**	3.37	33
v12	9	648	71/1	**2.549**	**8.10**	**65**	**83**	3.49	37
v13		649	71/1	**3.173**	**9.74**	**76**	**80**	3.49	10
v14	10	Inv.11578	71/1	**2.779**	**9.42**	**70**	**76**	3.14	13
v15		Inv.11579	71/1	**2.977**	**9.17**	**68**	**76**	3.45	16
v16		Inv.11573	89/1	**3.218**	**9.49**	**71**	**77**	3.61	13
v17		Inv.11574	91/1b	**2.829**	**9.06**	**67**	**76**	3.33	18
v18		3744	92/1	**2.905**	**8.69**	**65**	**78**	3.60	24
v19	14	675	93/1	**2.660**	**8.07**	**65**	**83**	3.67	38
v20		5058	95/1	**3.000**	**9.24**	**68**	**76**	3.45	15
v21		Inv.11580	95/1	**2.710**	**9.11**	**67**	**76**	3.17	17
v22		Inv.11581	95/1	**2.485**	**8.18**	**65**	**82**	3.36	35
v23		5086	95/2	**1.182**	**8.66**	**65**	**78**	1.47	25
v24	17	666	97/1	**2.894**	**9.09**	**67**	**76**	3.39	17
v25		670	98A/1	**2.385**	**8.08**	**65**	**83**	3.28	38
v26		671	98A/1	**3.021**	**9.27**	**69**	**76**	3.47	15
v27	18	672	98A/1	**2.187**	**7.64**	**65**	**87**	3.25	49
v28	19	601	102/1	**2.826**	**8.55**	**65**	**79**	3.58	27
v29		602	102/1	**2.322**	**8.43**	**65**	**80**	3.01	29
v30		603	102/1	**3.031**	**8.90**	**66**	**76**	3.64	20

Table 3: Data of the investigated victoriati of the Museo Archeologico di Torino, with their inventory or Fabretti numbers (Fabretti 1881). The column A is a catalog number for these victoriati. Column B indicates if the coin is comprised in the catalog of Table 1, providing the corresponding catalog number. Museum references and *RRC* type (Crawford, 1974) follow in column C and D. Weight, SG, core and average fineness are reported in columns E, F, G and H respectively. In columns I and L the original flan weight and the corresponding weight loss is reported.